



Uncertainty Quantification
Methodology for Calculation of IMSR Off-
Gas Source Term
Technical Report

Revision 2

February 2024

Abstract

This document presents a methodology for performing uncertainty quantification analysis for the Terrestrial Energy Integral Molten Salt Reactor (IMSR) source term based on coupled modeling of neutronics, thermal hydraulics, nuclide mass transport, and chemical speciation. The result of this effort will be a general, coupled, modeling capability for MSR systems, including the capability to propagate uncertainties in model input parameters for purposes of mass accountancy and the calculation of source term. The capability will be applicable to the fluoride fuel salt of the IMSR and will be useful in the analysis of off-gas release during steady-state and accident scenarios. In addition, these tools also provide a high-fidelity benchmarking opportunity for the vendor and the regulator to use as a reference tool for future licensing submittals when appropriate, especially when test and operational data are not abundantly available (e.g., new methods and new design configurations).

Non-Proprietary

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ABBREVIATIONS

CALPHAD	CALculation of PHAse Diagrams
ISMR	Integral Molten Salt Reactor
MCMC	Markov Chain Monte Carlo
MSBR	molten salt breeder reactor
MSR	molten salt reactor
MSRE	Molten Salt Reactor Experiment
NEAMS	Nuclear Energy Advanced Modeling and Simulations
ORNL	Oak Ridge National Laboratory
QoIs	quantities of interest
UQ	uncertainty quantification

NOTATION

v_{ll}	liquid velocity
v_{gg}	gas velocity
ϕ	neutron flux
σ	microscopic cross section
Σ_{ff}	macroscopic fission cross section
Σ_{aa}	macroscopic absorption cross section
β	delayed neutron fraction
$\beta\beta_{pp}$	delayed neutron fraction for precursor group p
ν	neutron per fission
V	neutron speed
k_{eff}	effective multiplication factor
λ_{pp}	decay constant for precursor group p
γ_j	species j yield
λ_{ij}	species j decay constant
D	neutron diffusion coefficient
P	pressure
T	temperature
A	flow area
g	gravitational acceleration

u_{ll}	bulk liquid internal energy
ρ_{ll}	bulk liquid density
FF'	frictional pressure drop per unit length
PP'	pressure increase per unit length due to external sources (i.e., pumps)
qq'''	volumetric heat generation rate
qq''	wall heat flux
μ_{ll}	dynamic viscosity
QQ	volumetric flow rate
LL	characteristic length
h_{ll}	convective heat transfer coefficient
kk_{ll}	thermal conductivity
cc_{pp}	specific heat capacity
CC_{pp}	concentration for precursor group p
CG_j^{ll}	isotope j concentration in bulk liquid phase
CG_j^{ww}	isotope j concentration at wall (deposition and erosion surfaces)
CG_j^{ss}	isotope j concentration at leaching surface
CG_j^{bb}	isotope j concentration at bubbles
γ_{jj}	fission yield fraction for isotope j
λ_{ij}	decay constant for isotope j
σ_{ij}	absorption cross section for isotope j
$\zeta_{ii \rightarrow jj}$	fraction of isotope i neutron capture producing isotope j
$\xi_{ii \rightarrow jj}$	fraction of isotope i decay producing isotope j
D_{ij}^{ll}	liquid diffusivity
Φ_{jj}	species j, molar flux at gas-liquid interface
aa_{jj}	species j, gas-liquid interfacial area
kk_{ij}^{bb}	mass transfer coefficient for bubbles
kk_{ij}^{dd}	mass transfer coefficient for deposition
$kk_j^{\text{le}}^{\text{le}}$	mass transfer coefficient for leaching
$kk_{ij}^{\text{er}}^{\text{er}}$	mass transfer coefficient for erosion

aa_{bb}	bubbles interfacial area for bubbles
aa_{ww}	wall interfacial area for deposition and erosion
aa_{ss}	interfacial area for leaching
cc_{jj}^{gg}	bulk gas concentration
DD_{jj}^{gg}	gas diffusivity in liquid
pp_{jj}^{gg}	bulk gas phase, species j partial pressure
kk_{jj}^{ii}	gas phase, species j partial pressure at interface
pp_{jj}^*	gas phase, species j theoretical partial pressure in equilibrium with cc_{jj}^{ll}
cc_{jj}^{ll}	bulk liquid phase, species j concentration
cc_{jj}^{ii}	liquid phase, species j concentration at interface
cc_{jj}^*	liquid phase, species j theoretical concentration in equilibrium with pp_{jj}^{gg}
TT_{gg}	vapor temperature
cc_{jj}^{gg}	species j, vapor molar density
$\rho\rho_{gg}$	vapor density
uu_{gg}	vapor internal energy
h_{gg}	vapor convective heat transfer
MM_{jj}	molecular weight of species j
MM_{ll}	molecular weight of bulk liquid
VV_{jj}	sum of the diffusion volume, species j
VV_{ll}	sum of the diffusion volume, bulk liquid

I. Purpose

The goal of this project is the development of a broadly applicable modeling and simulation capability and methodology for the propagation of uncertainties in model parameters of importance to molten salt reactor (MSR) source terms and off-gas systems. Oak Ridge National Laboratory (ORNL) will then use details of the specific fuel and coolant conditions of the (IMSR) being developed by Terrestrial Energy USA (TEUSA) to demonstrate the capabilities of the methodology in calculation source terms that could be supportive of future licensing submittals. Uncertainties to be addressed include those related to nuclear data (e.g., cross sections and yields), the physical properties of the salt (e.g., solubilities and viscosities), thermochemical properties (e.g., vapor pressures), and system design parameters such as power level and fluid flow rates. The developed capability will provide the boundary conditions to off-gas systems and will be used for the determination of the source term for postulated radioactive releases important to the licensing analyses.

The scope of the source term model development effort is to calculate the source term for a postulated accident design basis accident. The design basis accident selected is a [[

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develop a methodology that can evaluate every possible failure mode of the IMSR or every potential release mode.

Uncertainties to be addressed include those related to nuclear data (e.g., cross sections and yields), the physical properties of the salt (e.g., solubilities and viscosities), thermochemical properties (e.g., vapor pressures), and system design parameters such as power level and fluid flow rates. The developed capability will provide the boundary conditions to off-gas systems and will be used for the determination of the source term for postulated radioactive releases important to the licensing analyses.

Off-gas properties/aspects to investigate include the following: [[

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work will leverage codes under active development within Nuclear Energy Advanced Modeling and Simulations (NEAMS) Program of the US Department of Energy (DOE) and the NEAMS Molten Salt Thermal Properties Database. The codes that will be leveraged include:

1. Shift, for neutronics, nuclide transmutation, and nuclear cross-section uncertainty;
2. SAM for fluid transport;
3. Mole for time-dependent species transport,
4. Scale for the library of cross sections, isotopic yields, and isotope decay constants.
5. Gibbs Energy Minimizer for equilibrium phase concentrations and chemical potentials; and
6. PYCALPHAD for the thermodynamic engine for ESPEI.

A primary goal of this work will be the development and exercise of the integrated capability for determining the composition and activities with uncertainties for source term and the off-gas system. This will include the propagation of uncertainties in model input parameters to establish requirements for a given off-gas system, which will also serve the purpose of providing a bound on source term under postulated accident scenarios.

2. Nuclide Contributors to Off-Gas Source Term

The transport and retention of radionuclides within molten salt reactor systems is highly dependent on its chemical form within the fuel salt, otherwise known as its chemical speciation, which may be comprised of gas, liquid and/or solid phases. During postulated accident scenarios, the chemical speciation of a radionuclide, its production/destruction through fission and radioactive decay will determine its transport and potential release to the environment. Chemical speciation for a given radionuclide depends on a number of critical factors including fuel salt chemical composition, fuel salt redox potential, radionuclide concentration, pressure, and temperature. Experience with the Molten Salt Reactor Experiment (MSRE) led to the characterization of fission and transmutation products as noble gases, salt-seeking elements, and noble metals (Kedl 1972). The noble gases of primary interest are Kr and Xe, which are insoluble in salt and readily form a gas phase (bubble formation). In contrast, salt seekers create non-volatile, low-volatile, and redox-dependent, low-volatile fluorides within the fuel salt.

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Table 1 displays an example list of fluoride salts and their boiling temperature that were of interest to the MSRE fuel handling. These are provided for illustrative purposes because of their low boiling temperature relative to the higher operating temperature of the TEUSA IMSR. [[

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Table 1. Boiling Points of Fluoride Salts (Lindauer 1965)

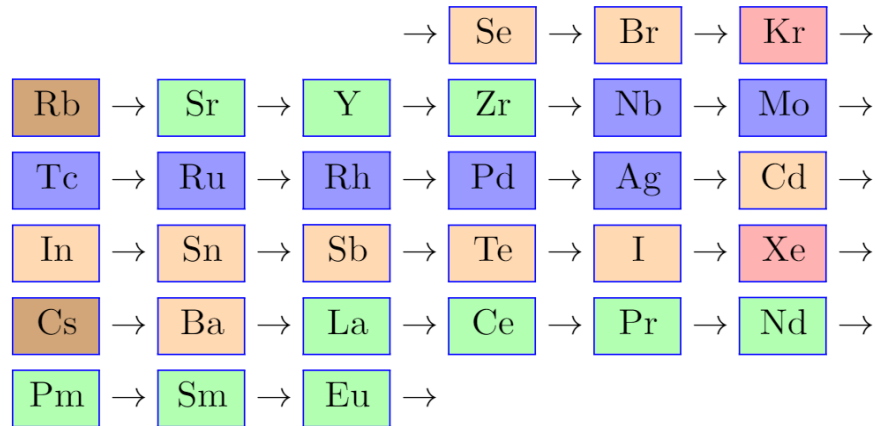
Salt	Boiling Temperature (°C)
CF ₄	-127.8
TeF ₆	-37.6
IF ₇	4.8
MoF ₆	38.9
UF ₆	56.5
Te ₂ F ₁₀	58.9
PuF ₆	62.0
IF ₅	102.0
CrF ₅	117.0
SbF ₅	149.5
MoF ₅	212.0
NbF ₅	236.0
RuF ₅	227.0
TeF ₄	283.9
SbF ₃	376.0
CrF ₄	400.0
RuF ₄	200.0

2.1 Radionuclides Assessment and Mass Accountancy

Figure 1 displays the nuclide chains that need to be assessed for source term development. Arrows from left to right within a row indicate nuclide transmutation with the progression continuing across subsequent rows. For example, the transmutation of Kr will result in Rb that, in turn, transmutes to Sr, and so on. Similarly, I will produce Xe, Cs, and Ba. Nuclides may be produced directly in fission, through radioactive decay, and through neutron capture. As indicated in Figure 1, the chemical behavior for each nuclide is quite different and includes noble gas, nonvolatile metals, and low-volatile fluorides. The chemical speciation depends on the chemical thermodynamic behavior of a given nuclide within the salt. [[

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Figure 1. Low-volatile fluoride (brown), Low-volatile fluoride with redox (orange), Nonvolatile fluoride (green), Nonvolatile metal (blue), and Noble gas (red)



Radionuclides are color coded by noble gas, noble metal, and the affinity for a given nuclide to create fluoride compounds (non-volatile and low-volatile fluorides). As shown, [[

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Table 2 displays the radionuclides requiring assessment for source term ranked by radiotoxicity. Dose factor is provided in units of millirems/millicurie. Also shown are the half-life for each nuclide (days), the estimated yield from ²³⁵U fission, and an initial characterization of the nuclide within a fluoride salt system based on MSRE experience. The initial characterization shows nuclides that may be categorized as noble gas, nonvolatile fluoride, low-volatile fluoride, and non-volatile (noble) metal using the same color coding as Figure 1. The low-volatile fluoride (redox dep) indicates nuclides with a dependence on redox potential. The column, label “MSRE system state”, shows an expected ordering from highest to lowest concentration for the given nuclide within the system state (salt, off-gas, and/or graphite/metal). Parentheses indicate a trace amount found in within the MSRE system. For example, [[

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III. Governing Equations for Molten Salt Thermal Fluid

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3.1 Frictional Losses

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3.2 Heat Generation and Removal

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3.3 Salt Thermophysical Properties and Fluid Closures

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IV. Governing Equations for Molten Salt Neutronics

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V. Governing Equations for Molten Salt Species Transport

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5.1 Bulk Liquid Phase Species Transport

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5.2 Gas-Phase Species Transport

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VI. Interphase Mass Transfer

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6.1 Correlation for Mass Transfer Coefficient

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6.2 Liquid Diffusivity

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6.3 Gas Diffusivity (in liquid)

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6.4 Henry's Gas Law Constant

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6.5 Gas Velocity

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6.6 Gas-Liquid Interfacial Area

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VII. Governing Equations for Plenum Gas Transport

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$$c_{jj}^{gg} = \frac{p_j}{p} \left(\frac{M_j}{M} \right)^{-1}$$

$$v_{gg} = \frac{p}{\rho_{gg}}$$

Species j molar density is related to the pressure, vapor temperature, and partial pressures:

where

$$P_{Pj} = \frac{p_j}{p} \left(\frac{M_j}{M} \right)$$

Vapor energy conservation for the gas mixture is analogous to Eq. 3:

Closure relations include the following:

where

$$\rho_{gg} = \frac{p}{R T} \quad (\text{kg/m}^3)$$

$$u_{gg} = \frac{p}{\rho_{gg}} \quad (\text{J/kg})$$

$$h_{gg} = \frac{p}{\rho_{gg}} \left(\frac{1}{R} \ln \left(\frac{p}{p_0} \right) + \frac{1}{\gamma} \right) \quad (\text{J/(mm}^2 \cdot \text{ss} \cdot \text{°K)})$$

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7.1 Gas Diffusivity (Plenum)

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VIII. Uncertainty Quantification Methodology Based on Governing Equations

8.1 Neutronics Analysis

The nuclide inventory for the actinides (U, Np, Pu, Cm, and Am) and fission products of Table 1 is determined by the solution of the steady-state eigenvalue equation for k_{eff} and the neutron flux ϕ of Eqs. 21 and 22, followed by solution of the coupled set of nuclide transmutation equations represented by Eq. 20 (Hightower 1975).

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8.2 Neutronics and Thermal-Hydraulics Uncertainty Propagation

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8.3 Chemical Speciation Uncertainty Propagation

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8.4 Uncertainty Propagation in Bulk Liquid - and Vapor - Phase Species Transport

Equations 24 and 25 represent species transport in the bulk liquid phase and gas phase, respectively, for nuclides defined in Table 1. For the bulk liquid, [[

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8.5 Dakota UQ Analysis

Table 3 summarizes the physics, model parameter input, and calculated QoIs based on the governing equations. [[
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The Dakota software from Sandia National Laboratories (Adams et al. 2021) provides a toolbox of advanced parametric analysis techniques enabling quantification of margins and uncertainty, risk analysis, model calibration, and design exploration with computational models. Dakota includes

methods for optimization, UQ, parameter estimation, and sensitivity analysis, [[]]

Table 3. UQ analysis model input and output Qols

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Random sampling methods are the “black-box” UQ work horse provided in Dakota (as well as the Sampler code used with Shift). Essentially, [[

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IX. Conclusions

The analyses and UQ methodology for determining off-gas nuclide composition based on the governing equations for coupled neutronics, thermal fluids, mass transfer, and chemical speciation has been established. A number of closure relations outlined in the document will be subject to change but will generally not impact the approach outlined.

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